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PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application : Cornelis Roeland Bayense et al.  
Application No. : 09/830,907  
Filed : June 19, 2001  
Confirmation No. : 5302  
For : STAR SHAPED ALUMINA EXTRUDATES AND  
CATALYST BASED THEREON  
Examiner : Edward M. Johnson  
Attorney's Docket : VER-177XX

TC Art Unit: 1754

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I hereby certify that this correspondence is being deposited with  
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Alexandria, VA 22313-1450 on 7-27-7.

By 

Charles L. Gagnebin III  
Registration No. 25,467  
Attorney for Applicant

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DECLARATION OF CORNELIS ROELAND BAYENSE UNDER 37 C.F.R. §1.132

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, Cornelis Roeland Bayense, hereby declare that:

1. I am an inventor of the above-referenced U.S. Patent Application Serial No. 09/830,907, filed on June 19, 2001 (the present application).

2. I hold a doctoral degree in Chemistry from the Eindhoven University of Technology, Eindhoven, NL. I currently hold the position of Senior Chemist in the Process Catalysis Research group at BASF Nederland BV, P.O. Box 19, 3453 ZG De Meern.

3. I am familiar with the prosecution history of the above-referenced U.S. patent application, including the Office Action mailed November 2, 2006 (the Office Action), which includes the rejection of claims 1-8, 10-16, and 20-21 for alleged obviousness over Mulaskey et al., U.S. Patent 4,554,268 (Mulaskey) in view of Neel et al., U.S. Patent 4,554,268.

4. In the Office Action, the Examiner has taken the position that Mulaskey suggests an extruded alumina having 0.05 ml/g of pore volume in pores of diameter greater than 10,000 Å (1000 nm).

5. I am familiar with the production of extruded alumina catalyst materials and protocols for testing their pore size distribution. I have supervised the preparation of extruded alumina materials according to Mulaskey (Mulaskey material) and according to the present application (BASF material). I have also

supervised the measurement of the pore size distributions of both materials.

6. The Mulaskey material was made by mixing aluminum trihydrate (Condea SB) with aqueous  $\text{HNO}_3$ , thus forming an alumina gel composition. The semi-dry compound was subsequently extruded using a one-screw extruder. The extrudates were dried at  $105^\circ\text{C}$  and subsequently calcined at  $540^\circ\text{C}$ . The resulting extrudates had a surface area of  $305 \text{ m}^2/\text{g}$ .

7. Two samples of the BASF material were prepared. One sample was made according to the procedure described in the EXAMPLE of the present application. 1.5 kg of aluminum trihydrate (0.98 g  $\text{Al}_2\text{O}_3$ ) with an average particle size of 30-50 $\mu\text{m}$  was mixed with 0.4 kg of alumina binder. The powders were mixed extensively while slowly adding aqueous  $\text{HNO}_3$  in an amount of 2 wt% (based on the total  $\text{Al}_2\text{O}_3$  weight). The alumina binder was peptized, and mixing was continued until a relatively dry product was obtained. The semi-dry mix was extruded using a one-screw extruder, equipped with a die having star-shaped holes and a cutting device. After drying the extrudates at  $105^\circ\text{C}$  for 16 hours, calcination was performed at  $850^\circ$ . The resulting star-shaped alumina catalyst had a typical surface area of approx.  $100 \text{ m}^2/\text{g}$ . The second sample of BASF material was a commercial sample.

8. The pore size distributions of the alumina catalyst samples were measured by Hg-porosimetry on a Thermo Finigan apparatus (Pascal 140/440), operating at pressures up to 400 MPa, corresponding to pore sizes down to 36 Å. For filling pores with mercury under pressure, the relation between pressure and pore size is expressed in the so-called Washburn equation, which is described below. The Washburn-equation is valid for cylindrical pores, open at both ends, where no changes occur to the surface tension of Hg or to the morphology of the sample during the experiment.

9. The Washburn equation is  $P * r = -2 * \gamma \cos \theta$ , where  $P$  = pressure,  $r$  = pore radius,  $\gamma$  = surface tension of Hg, and  $\theta$  = contact angle (on average 141.3°).

10. For the measurement, a few grams of extruded alumina sample (typically corresponding to 0.25 ml of expected total pore volume) was loaded into a sample holder (dilatometer), which was mounted onto the apparatus. The sample was subsequently put under vacuum (0.01 kPa). In the next step, the dilatometer was filled with Hg, which gave the total Hg-pore volume. Then the pressure in the dilatometer was increased to 400 MPa, and as the pressure was increased, more and smaller pores were filled with mercury. The amounts of mercury penetrating into the sample at increasing mercury pressures were processed using the Pascal 140/440 software

(using the Washburn equation) and thus translated into a pore-size distribution graph. The results are shown in attached Table 1 and Figure 1.

11. The Mulaskey material was found to have a total pore volume of 0.4 ml/g and showed a pore size distribution concentrated in the range below 1,000 Å (100 nm). No pores were observed having diameters greater than 10,000 Å (1,000 nm). See Table 1 and Fig. 1, filled bars.

12. The BASF material obtained from a commercial sample was found to have a total pore volume of 0.44 ml/g, with 0.07 ml/g of pore volume in pores larger than 10,000 Å (1,000 nm) diameter. See Table 1 and Fig. 1, empty bars.

13. The BASF material prepared according to the EXAMPLE of the present application was found to have a total pore volume of 0.56 ml/g, with 0.07 ml/g of pore volume in pores larger than 10,000 Å (1,000 nm) diameter. See Table 1. Although the complete pore size distribution data are not shown for this sample, I would expect it to show a pore size distribution very similar to that shown in Fig. 1 for the BASF commercial sample.

14. The pore distribution results described above demonstrate that no measurable pore volume was found in pores of diameter greater than 10,000 Å (1,000 nm) for the Mulaskey material. For both samples of the BASF material, however, 0.07

ml/g of pore volume was present in pores of diameter greater than 10,000 Å (1,000 nm), consistent with the claimed range.

15. Therefore, in my opinion, an extruded alumina prepared according to the disclosure of Mulaskey has far less than the value of 0.05 ml/g of pore volume in pores of diameter greater than 10,000 Å (1,000 nm) which is required by the present claims. A pore volume of 0.05 ml/g in pores of diameter greater than 10,000 Å (1,000 nm) easily would have been detectable by the Hg porosimetry assay performed in this study; however, no pore volume was detected in that range for the Mulaskey material.

I hereby declare that all statements made herein on personal knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 11<sup>th</sup> day of July, 2007.

By

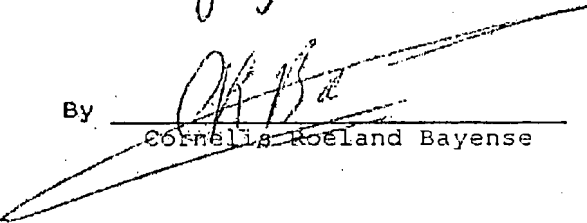
  
Cornelis Roeland Bayense

Table 1

	Mulaskey Al <sub>2</sub> O <sub>3</sub> (US Patent 3,673,079, 1972)	BASF Starshape Al <sub>2</sub> O <sub>3</sub> (US Pat. Appl. Nr. 09/830,907, Example 1)	BASF Starshape Al <sub>2</sub> O <sub>3</sub> (US Pat. Appl. Nr. 09/830,907, Commercial catalyst)
BET-S.A. (m <sup>2</sup> /g)	> 300	106	96
Total-PV (ml/g)	0.25-0.40	0.56	0.44
Macro-PV (ml/g)*	0	0.07	0.07
MPV/TPV (%)	0 (max. 10%)	12.5	15.9
SCS (N)	no data	65	64
BCS (MPa)	no data	1.08	1.08

\* MPV = Macro-PV = PV in pores with diameter >10,000Å